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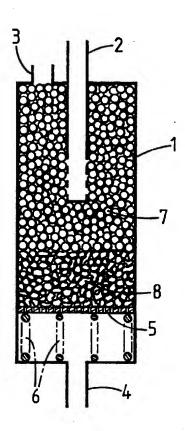
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(54) Title: APPARATUS AND PROCESS FOR VAPOUR RECOVERY

(57) Abstract

A canister (1) for attachment to a vehicle fuel tank to reduce hydrocarbon emissions comprises a gas inlet (2), gas outlet (3) and vent (4) and contains a bed of adsorbent polymer (7) and a bed of carbon (8). The vent opens into the carbon bed and the gas inlet and outlet open into the polymer bed.



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⁺ It is not yet known for which States of the former Soviet Union any designation of the Soviet Union has effect.

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APPARATUS AND PROCESS FOR VAPOUR RECOVERY

The present invention relates to the recovery of gasoline vapours from mixtures of gasoline vapour and air.

Gasoline vapour is emitted from motor vehicles powered by gasoline engines as a consequence of displacing gasoline vapour from the fuel tank during refuelling (refuelling losses). It is also emitted as a consequence of evaporation from the engine and fuel system either when the vehicle is in operation or when standing after use (diurnal losses). The emission of gasoline vapour is considered to be undesirable.

Manufacturers already fit carbon canisters to eliminate the diurnal losses and such an arrangement is disclosed for instance in GB 1 416 336. The carbon adsorbs the vapour emitted when the engine is stationary after use. The carbon canister is then regenerated by drawing air through the canister using the vacuum generated in the engine inlet manifold so that the gasoline vapour recovered from the canister is mixed with the normal air/fuel mixture to the engine. The vapour emission when the vehicle is stationary is quite small so that a canister having a capacity of about 1 litre is thought to be satisfactory. During normal vehicle operation the vapours emitted are continuously returned to the engine.

Refuelling an almost empty tank involves displacing a large amount of gas (corresponding to the volume of the fuel tank if it is being completely refilled) saturated with gasoline vapour although this only occurs approximately once every 600 km. This is equivalent to approximately 150-200 g of vapour for a typical

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European car (60 L tank) depending on the composition of the vapour. There are proposals both in the US and in Europe to impose legal restrictions on the amount of gasoline vapour which may be released into the atmosphere during refueling. One method proposed to meet such legal requirements is the recycling of the vapour from the fuel tank connected to the engine back to the storage tank from which the vehicle is refueled. This however is an expensive procedure in view of the complex equipment needed, and the work required at numerous refueling stations, often with underground storage tanks. It would be desirable to find a simple method of dealing with the problem by an extension of the use of adsorbent canisters mounted in vehicles. There is however a problem in trying to eliminate gasoline vapour emission during refuelling by the use of canisters carried in the vehicle. Although the overall weight of the gasoline vapour to be adsorbed is quite small the requirement to trap this during the 2-3 minutes that it takes to fill a normal saloon car tank would make it necessary to use as much as 5 litres of the carbons currently in use. Such large quantities of carbon can not be conveniently be provided in the restricted engine

One approach to reducing the size of the canister is to use a carbon with a high adsorptive capacity. However we have found that such carbons are not easily regenerated in use by drawing air through the carbon bed. They would adsorb a large quantity of gasoline vapour on the first cycle, but only small quantities thereafter. The optimum active carbon the adsorption of gasoline vapours is one which has a high pore volume with pores in the small mesopore range (approximately 2 nm diameter). This maximises both the adsorption capacity of the carbon and the regenerability resulting in the largest possible working capacity. However the working capacity for typical gasolines is still limited to approximately 5% weight predominantly by the difficulty of regenerating the bed.

compartment of modern saloon cars.

GB 1 416 336 discusses some of the disadvantages of the use of carbon and proposes to overcome them by replacing the carbon by a

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particulate macroreticular, substantially non-ionogenic, water-insoluble polymer having a specified surface area, porosity and average pore diameter. Among the polymers which may be used are polymers of divinyl benzene.

However we have found that such porous polymers are not satisfactory for use as gasoline adsorbents. The porous polymers have a good saturation capacity for gasoline vapour and are easily regenerated. However the lower molecular weight components of the gasoline vapour, such as propane and butane, are not held strongly enough on the polymer and thus break through the adsorbent bed and escape into the atmosphere after a comparatively short time thereby restricting the working capacity of the bed.

In principle it is always preferably to prepare an adsorbent from a single material so as to simplify the manufacturing process. We have however found that the use of two different adsorbent materials give sufficiently marked advantages to justify the increased complexity.

According to the present invention a canister for attachment to a vehicle fuel tank, said canister comprising a body adapted to receive an adsorbent, and having a gas inlet adapted to be connected to a gasoline storage tank, a gas outlet adapted to be connected to a gasoline engine, and a vent to the atmosphere, is characterised in that the canister contains a bed of adsorbent polymer and a bed of carbon arranged such that the vent opens into the bed of carbon and the gas inlet and gas outlet open into the bed of adsorbent polymer.

The present invention further provides a motor vehicle with a gasoline engine having a canister defined as above.

According to the present invention a method of adsorbing gasoline vapour from air comprises passing the mixture of air and gasoline vapour through a first bed of adsorbent polymer, and then through a bed of adsorbent carbon, and subsequently regenerating the adsorbent beds by passing a stripping gas first through the bed of adsorbent carbon and then through the bed of adsorbent polymer.

According to another aspect of the present invention a process for operating a gasoline engine of a motor vehicle comprises passing

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air displaced from the fuel tank while refueling the vehicle through a first bed of adsorbent polymer, and then through a bed of an adsorbent carbon so as to adsorb high molecular weight components of the gasoline on the polymer and lower molecular weight components of the gasoline on the carbon, and subsequently operating the engine so as to draw air though the bed of adsorbent carbon and then through the bed of absorbent polymer before feeding the air to the engine.

The mixture of air and desorbed gasoline components will generally be blended will the normal gasoline/air fuel before the total mixture is passed to the engine.

The adsorbent carbon may be any of the conventional active carbons already proposed for use in gasoline engines. However the use of the carbon in conjunction with the polymer adsorbent bed in the layered bed canister allows the use of a wider variety of active carbons where the requirement for the majority of pores to be small mesopores can be relaxed. Using conventional premium European gasoline the best results in the layered bed canister have been obtained with a highly microporous coconut shell carbon.

Polymers which are suitable adsorbent polymers for gasoline vapour adsorption are disclosed in GB 1 416 336. This discloses the use of particulate macroreticular, substantially non-ionogenic, water-insoluble polymers having a specific surface area in the range-10 to 10000 m²/g, a porosity of 25% to 85%, and an average pore diameter of 2 nm to 2000 nm. Among specific polymers which may be used are those obtained by polymerising under macroreticular polymer producing conditions. The monomer charge is composed of ethylenically unsaturated monomer or monomers and containing from 2% to 100% by weight, based on the weight of the charge, of 1 or more of divinyl benzene, trivinyl benzene, alkyl vinyl benzenes having from 1 to 4 methyl or ethyl groups substituted in the benzene nucleus, and alkyl divinyl benzenes having from 1 to 3 methyl or ethyl groups substituted in the benzene nucleus.

We believe that the surface area of the polymer is preferably in the range 200 to 2000 $\rm m^2/g$, and that the polymer should contain a significant volume of pores in the range 1 nm to 50 nm for

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adsorption purposes, as well as larger pores for gas transfer.

A particularly preferred polymer is a copolymer of a mixture containing a major amount of divinyl benzene and a minor amount of ethyl styrene which is sold under the designation "Ambersorb XAD4" by Rohm & Haas Inc.

The quantities of adsorbent used will depend on the working adsorbing capacity of the polymer, the working adsorbing capacity of the active carbon, the quantity of gasoline vapour to be adsorbed between each regeneration period and the time available for regeneration. For use in a canister useable in a normal size family car the total volume of adsorbent may be in the range 500 ml to 5 litres.

The optimum amount of the two different adsorbents to be used will depend on the design of the canister. The designated ratios refer to the volume of adsorbent between the gasoline vapour inlet and the vent to atmosphere. This can be seen by reference to figures 1 and 2. In figure 1 which shows a canister with the vapour inlet (2) in the centre of the bed the polymer:carbon ratio refers to the volume of adsorbent between the vapour inlet (2) and the vent to atmosphere (4). In the canister shown in figure 2 the ratio refers to the total volume of adsorbent in the canister between the vapour inlet (12) and the vent to atmosphere (14). The optimum ratio of polymer to carbon will vary with the type of gasoline to be used but may be in the range 3:7 to 7:3, or preferably in the range 4:6 to 6:4, more preferably 4.5:5.5 to 5.5:4.5.

The relative volumes of the two adsorbents may be adjusted to optimise the performance of the canister system for different types of gasoline used in different regions of the world. For the higher volatility fuels typically used in Europe a volume ratio of polymer to carbon of about 1:1 is preferred. For lower volatility fuels it is believed that polymer to carbon volume ratios of greater than 1:1 (ie larger relative amounts of polymer) will be desirable.

Depending on the design of the canister and the disposition of the inlet and vent, the ratios described above may correspond to a canister having a relatively large bed of adsorbent polymer and a

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relatively small bed of carbon. However this will not necessarily correspond to the most efficient use of the adsorbents.

The invention will now be illustrated with reference to the drawings and the following experiments.

In the drawings:

Figure 1 is a diagramatic cross-sectional view of one form of a gasoline adsorbing canister for use in the present invention, and Figure 2 is a cross-sectional diagramatic view of an alternative form of gasoline canister according to the invention.

Referring to Figure 1 a cylindrical canister (1) is provided with a gas inlet (2) extending downwardly into the body of the canister and opening into the canister through slots at its lower end. A vapour outlet (3) is provided at the top of the canister. vent (4), which may allow air to both enter or leave the canister is provided at the lower end. A perforated plate (5) is mounted above springs (6) which serves to urge the plate (5) upwards so as to keep any adsorbent material in the upper part of the canister compressed into its packed state. The upper part of the canister between the connection to the vehicle inlet manifold (3) and the vapour inlet (2) is filled with a bed (7) of adsorbent polymer particles. lower part of the canister is filled with two layers. The upper layer (1) is of the porous polymeric adsorbent whilst the lower layer (8), closest to the atmospheric vent (4) is filled with carbon adsorbent such that the volume ratio of the volume of bed (1) to bed (8) is most preferably in the ratio 4.5:5.5 to 5.5:4.5.

In use in a motor vehicle the inlet (2) is connected by piping and appropriate valves to the vehicle's fuel tank connected by piping and appropriate valves to the vehicles fuel tank and to the vents from the vehicles fuel inlet system and crankcase. The outlet (3) is connected by piping and an appropriate valve to the engine, for example it may be connected as a bleed to a carburettor. The vent (4) provides for the inlet or outlet of gas to the outside air. When the motor vehicle is refueled gas displaced from the fuel tank enters the canister through inlet (2). Outlet (3) can be closed so that no air is drawn through it to the engine but this is

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not essential. Gasoline vapour evaporating from the fuel tank will pass through inlet (2) into the canister. It will initially contact the absorbent polymer and then diffuse through this into the adsorbent carbon. When the motor is running air will be drawn in through the vent (4) and will sweep adsorbed gasoline vapour components out of the adsorbent carbon. The air will then pass through the adsorbent polymer to the engine by way of outlet (3) so regenerating the adsorbent materials in the canister.

Figure 2 shows an alternative design of canister for use particularly in reducing emissions during the refueling of motor vehicles. The canister (11) is provided with a gas inlet (12) and a gas outlet (13) together with a vent (14). These are all provided at the top of the canister. A metal divider (15) covered with an asbestos insulating sheet (16) divides canister (11) so that an elongated flow path is provided between the inlet (12) and outlet (13) on the one hand and the vent (14) on the other. The canister is filled with two types of adsorbent material, namely a bed of adsorbent polymer (17), and a similar bed of adsorbent carbon (18). Wire mesh or perforated plates (19) and glass fibre (20) hold the adsorbent particles in position.

In use air containing gasoline vapour displaced from a motor vehicle fuel tank during refueling passes into the canister through inlet (12) and passes through the bed of adsorbent polymer (17) and then the bed of adsorbent carbon (18) to the vent (14) through which it is discharged to the atmosphere. When the canister is to be regenerated the connection to the fuel tank can be closed by a valve (not shown) and a connection is opened to the engine through outlet (13) by which air will be drawn into the canister through vent (14) so as to displace adsorbed gasoline vapour from the adsorbent particles and to transport it through outlet (13) to the engine.

Tests were carried out on a Volkswagen car supplied fitted with

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a carbon adsorbent canister of the type shown in Figure 1 having a capacity of 1.1 litre and filled with a commercial particulate carbon adsorbent. The engine was subjected to cycles of (a) running, in which air was drawn through the canister into the engine and (b) standing, in which gasoline vapour from the fuel tank passed through the canister before air from the tank was vented to the atmosphere. After use for a considerable number of cycles so as to give a fully aged carbon, tests were carried out over six cycles to determine the working capacity for gasoline adsorption of the carbon. This was done by measuring weight increase during the period of gasoline adsorption. The result is shown in the Table. Comparative Test B

An experiment was carried out as in Comparative Test A except that the carbon particles were removed from the canister which was then loaded with a gasoline adsorbing polymer (1.1 litre packed volume) which was a polymer of divinyl benzene containing some ethyl styrene sold under the commercial designation "Ambersorb XAD4".

The working capacity of the polymer was determined as in Comparative Test A.

20 Example 1

An experiment was carried out as in Comparative Test B, except that the lower part of the canister connected to the fuel tank and the air feed to the engine was loaded with the carbon of Test A, while the upper part was loaded with the adsorbent polymer of Test B. Both adsorbents were fully aged.

The relative quantities of gasoline-adsorbing polymer and absorbent carbon were 3:1 by packed volume.

The working capacity of the mixture was determined as the mean of six cycles of the operation with a standard deviation of 3.54 g. The result is given in the Table.

For comparison a predicted value of the working capacity was determined based on the measured capacities from Tests A and B and the relative amounts of carbon and polymer used in example 1. The results are given in Table 1.

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TABLE

EXPERIMENT	WORKING CAPACITY	
Test A	20	
Test B	22	
Example 1	29.2	
Example 1 (predicted)	21.5	

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In addition to these tests with typical vehicle canisters, laboratory tests were carried out with the equipment described below. The two adsorbent beds (1 and 2) were contained in steel vessels that could be separated and weighed individually after each adsorption and desorption cycle. Each vessel was 4 cm in diameter and could hold up to 80 ml of adsorbent held in place between plugs of glass wool. The two beds were contained in a water bath maintained at 53C. The adsorption cycle was simulated by passing gasoline vapour, produced by bubbling 83 ml min of nitrogen through 200 ml Eurograde unleaded 95/85 gasoline held at 200C in a water bath, through the polymer bed and then the carbon bed in series. The temperature in the centre of each bed and the total flow from the second carbon bed was recorded as a function of time. The composition of the effluent gas was analysed by mass spectrometer. The adsorption cycle was assumed to be complete when the 29 peak in the mass spectrometer (mainly due to propane) reached 1×10^{-8} unts, ie partial breakthrough. The gas flow was then stopped and the two beds were isolated and weighed. The two beds were then returned to the water bath.

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Description was carried out by reversing the nitrogen flow through the two beds. An average flow rate of 485 ml/minutes was used for a fixed time of 40 minutes for the description cycle. After the 40 minutes the two beds were again removed from the water bath and weighed.

Comparative Tests C and D

Tests C and D (table 2) are not according to the present invention and show the breakthrough time in minutes and the working capacity in both % weight and g/litre for the tests where both vessels contained either 80 ml of the porous polymer, XAD4 (test C) or 80 mls of a typical extrudate active carbon recommended for use in evaporative loss canisters (test D).

Examples 2 to 7

in both the time to breakthrough and the working capacity when the first bed is filled with 80 ml of the porous polymer, XAD4, and the second bed with 80 ml of activated carbon. In Examples 2 the carbon is the recommended extrudate active carbon used in comparative test D. It can be seen from a comparison of tables 1 and 2 that the increase in the working capacity compared to both the polymer/polymer bed and the carbon/carbon beds is almost identical to that found in the vehicle tests whilst the actual working capacities are slightly higher for the laboratory test probably due to slight differences in the breakthrough criterion. This

Examples 3 to 7 demonstrate the use of different activated carbons in the second adsorbent bed. These materials were not selected using the normal criterion for evaporative emission canister carbons of maximising the small mesopore (2nm) volume but were typical microporous active carbons. It can be seen that the best perforance for the dual bed system was achieved with Sutcliffe Speakman AC610C, a highly microporous active carbon. The relative performance of the activated carbons in the dual bed system correlates better with the saturation butane capacity of the carbons than the working capacity which are also shown in table 2. These

results demonstrate a second benefit of the dual bed canister, namely that the carbon component can now be selected to maximise the butane adsorption capacity without the generally poorer regeneration capability of the microporous carbons influencing the working capacity.

Table 2 - Cyclic Performance of Laboratory Test - twin beds each of 80 ml capacity.

10	Test	Materials	Breakthrough time	Working (Capacity	Butane Ads	orption
10			(minutes)	% weight	g/litre	Saturation % Weight	Working g/L
*	c	XAD4/XAD4	21.8	7.0	27.6	32	98
15	С	AC1/AC1	26.0	6.8	29.1	33	63
	2	XAD4/AC45	40.5	11.0	44.8	39	54
	3	XAD4/610C	42.2	11.7	46.0	34	50
.	4	XAD4/GAC616G	38.0	9.2	42.2	30	81
	5	XAD4/AS IV	36.0	10.9	41.7	33	63
20	6	XAD4/WS IV	33.0	9.9	37.9	33	75

Comparative Test E and Examples 7 and 8

The same test conditions as used for tests C and D and Examples 2 to 6 were used in the following examples of a less preferred form of the present invention. In examples 2 to 6 the ratio of polymer to carbon was 50:50. In examples 7 to 8 the volume of adsorbent in the second bed has been reduced to 16 ml. The polymer:carbon volume ratio in tests F and G was 83:17.

Table 3

Test time	Materials (minutes)	Breakthrough % weight	Working Capacity
Е	XAD4/XAD4	10.8	5.3
7	XAD4/SS208C	12.5	5.9
8	XAD4/AC1	11.6	6.0

As the total bed volume has been reduced from 160 mls to 96 mls the breakthrough times and working capacities in table 3 cannot be compared directly with those shown in table 2.

Claims:

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- 1. A canister for attachment to a vehicle fuel tank, said canister comprising a body adapted to receive an adsorbent, and having a gas inlet adapted to be connected to a gasoline storage tank, a gas outlet adapted to be connected to a gasoline engine, and a vent to the atmosphere, is characterised in that the canister contains a bed of adsorbent polymer and a bed of carbon arranged such that the vent opens into the bed of carbon and the gas inlet and gas outlet open into the bed of adsorbent polymer.
- 2. A canister according to claim 1 wherein the volume ratio of adsorbent between the gasoline vapour inlet and the vent to atmosphere is in the range 3:7 to 7:3.
- 3. A canister according to Claim 2 wherein the volume ratio is in the range 4:5:5.5 to 5.5:4.5.
- 4. A canister according to claim 1 wherein the canister contains a relatively large bed of adsorbent polymer and a relatively small bed of carbon.
- 5. A canister according to anyone of the preceding claims wherein the adsorbent polymer is a particulate macroreticular substantially non-inorganic, water insoluble polymer having a specific surface area in the range 10 to 1000 mHg, a porosity of 25% to 85%, and an average pore diameter of 2 nm to 2000 nm.
- 6. A canister according to any one of the preceding claims wherein the polymer has a surface area in the range 200 to 2000 $\rm m^2/g$ and contains pores in the range 1 to 50 nm.
- 7. A canister according to any one of the preceding claims wherein the total volume of adsorbent is in the range 500 ml to 5 litres.

- 8. A motor vehicle with a gasoline engine equipped with a canister, which canister is according to any one of the preceding claims.
- 8. A method of adsorbing gasoline vapour from air comprises passing the mixture of air and gasoline vapour through a first bed of adsorbent polymer, and then through a bed of adsorbent carbon, and subsequently regenerating the adsorbent beds by passing a stripping gas first through the bed of adsorbent carbon and then through the bed of adsorbent polymer.
- 10. A process for operating a gasoline engine of a motor vehicle comprises passing air displaced from the fuel tank while refueling the vehicle through a first bed of adsorbent polymer, and then through a bed of an adsorbent carbon so as to adsorb high molecular weight components of the gasoline on the polymer and lower molecular weight components of the gasoline on the carbon, and subsequently operating the engine so as to draw air though the bed of adsorbent carbon and then through the bed of absorbent polymer before feeding the air to the engine.

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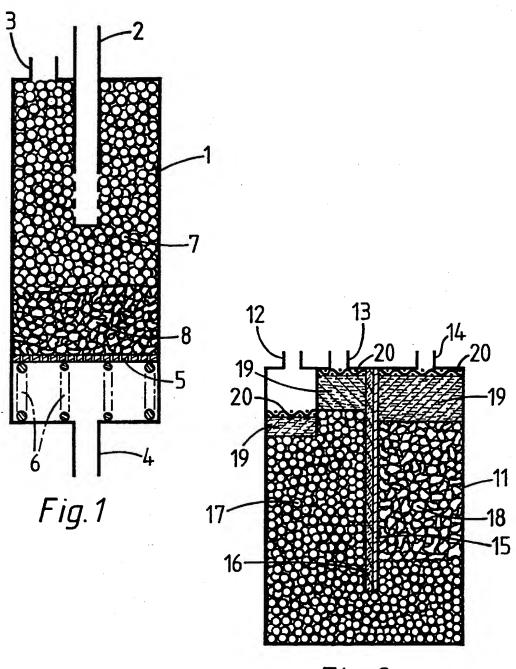


Fig. 2

INTERNATIONAL SEARCH REPORT

International Application No

PCT/GB 91/01269

I. CLASSIF	ICATION OF SUBJE	CT MATTER (If several classification symbol	ols apply, indicate all) ⁶	
According to		Classification (IPC) or to both National Classi B60K15/00 ; F02M25/08	ification and IPC	,
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ni. Docu	MENTS CONSIDERE	ED TO BE RELEVANT		Relevant to Claim No.13
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IV. CERI	TIFICATION			
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GB-A-1416336	03-12-75	US-A- 3805493 US-A- 3798876 CA-A- 990221 DE-A- 2258298 FR-A,B 2162081 JP-C- 979629 JP-A- 48063973 JP-B- 54012429	23-04-74 26-03-74 01-06-76 14-06-73 13-07-73 14-12-79 05-09-73 23-05-79
US-A-3838673	01-10-74	None	
US-A-3844739	29-10-74	None	
US-A-4289513	15-09-81	None	
DE-A-3842994	05-07-90	None None	